

Ultrafast switching of nanostructure in fused silica

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Abstract: Femtosecond switching of anisotropic nanostructure indicating birefringence with a time constant of ± 200 fs was observed. Such anisotropy has evolved by lowering threshold for defect formation and enhanced coherency of the electron plasma wave.

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Ultrashort pulse lasers have previously allowed probing of molecular dynamics in real time on the femtosecond time scale, especially exotic behavior ranging from alignment of molecules and clusters, structural deformation, Coulomb explosion, X-ray emission, to nuclear fusion has been well-researched. Recently, it has been reported ultrafast phenomena of phase transitions on solid [1], alignment of molecules in liquid [2], and modification of electron localization in magnetic materials [3], which are realized by not only electron-to-lattice coupling, but also interaction between light electric field and plasmon or magnetic dipole. Here we demonstrate experimentally a temporal evolution of optical anisotropy by femtosecond double pulse varying the delay and polarization of the two pulses. Such anisotropic modification is reversible and can occur with a time constant of ± 200 fs.

In the double pulse experiments, a series of dots were written in fused silica using a linearly polarized femtosecond double pulse with a different polarization direction varying the delay time (τ_{delay}) between the two pulses. A pair of polarization beam splitters was used to split and recombine the beam to create double pulse. One of the two beams was delayed by an optical delay and rotated the polarization in an orthogonal direction with respect to that of another beam. The double pulse was focused via a microscope objective at a depth of about 50 μm below the sample surface. The pulse energy of both pulses was about 1.0 μJ and the beam power measured after microscope objective was independent on the orientation of light polarization. For purpose of comparison to the common single beam experiment (Fig. 1a, 1b), one of the two beams was blocked for each beam and successively focused on the same location. After writing, the structures were inspected under an optical microscope and a polarized optical microscope.

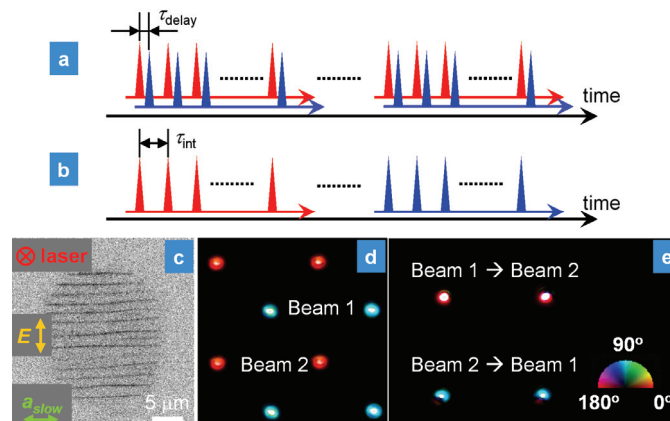


Fig. 1 Comparison of the slow axis of birefringence between the common single beam (a) and the double pulse (b) experiments. Anisotropic nanostructure consists of structural modulation with oxygen concentration in glass (c). The birefringence created by single beam (d) is rewritable (e).

Previously, we reported that the periodic nanostructure was self-organized in the orthogonal direction to the light polarization (Fig. 1b), which were interpreted in terms of the interference pattern between photon and plasmon in glass [4]. Such nanostructure consists of structural modulation with

oxygen concentration in glass indicates form birefringence, because the refractive index in the direction perpendicular to the light polarization is higher, i.e. the slow axis of birefringence (a_{slow}) is certainly perpendicular to the light polarization (E) (Fig. 1c). The polarization-dependent nanostructure can act as a bit of optical memory, in which has been naturally incorporated the angular resolution capability. The experimental results of the successive focusing of each single beam reveal that the slow axis with azimuth angle perpendicular to the light polarization can be reproduced, namely birefringence is rewritable (Fig. 1d). Fig. 2 shows time-resolved measurements for the azimuth angle of birefringence using double pulse configuration with the time resolution of 80 ± 40 fs where the azimuth angles observed by each single beam experiments were also shown. We observed the azimuth angle in double pulse experiments approached from initial as low as created the first pulse asymptotically to the intermediate value between that of the first and the second pulse ($= (a_{slow}^{2st} - a_{slow}^{1st})/2$) (Fig. 2b). This femtosecond modification of birefringence with the time scale of ± 200 fs could be explained by rotation of the direction of nanostructure created by the first pulse according to the second pulse with the orthogonal polarization direction to that of the first pulse. The discrepancy between the successive irradiation for each single beam with different polarization direction (Fig. 1e) and the double pulse irradiation with time delay (Fig. 2) is due to insufficient evolution of the nanostructure corresponding to the polarization direction of the second pulse. On the other hand, we can conclude that the birefringence can be well controlled by the polarization direction of only one pulse.

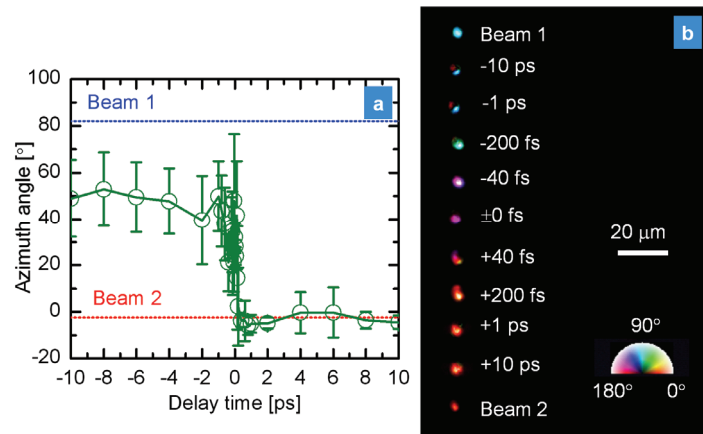


Fig. 2 Azimuth angle of birefringence for various delays of orthogonally polarized double pulses. Right images indicate the direction of slow axis observed by a polarization microscope.

In order to reveal the dynamics of the anisotropic nanostructure formation, we have also observed the evolution of birefringence varying the interpulse time and the number of pulse with the same peak power density of 1.0×10^{15} W/cm². From these experiments, we have observed typical three types of birefringence induced by single femtosecond pulses. Residual birefringence was characteristically observed at the irradiated pulses fewer than 100, which was originated from internal stress distribution due to instantaneous structural change with the formation of oxygen defects or heat and shock wave propagation. In the middle regime, the apparent form birefringence produced by the anisotropic nanostructure was observed in the center of the residual birefringence. Finally, the significant birefringence can be observed around the beam focus volume over thousands of pulses irradiation. These results indicate that at least one hundreds pulses are required for the anisotropic nanostructure formation, regardless of interpulse time. We have interpreted the evolution of optical anisotropy in glass created by femtosecond pulses in terms of lowering threshold for the oxygen defect formation and improving coherency of electron plasma wave. Indeed, homogeneous oxygen defects were formed only in the center of focal spot by the one pulse shot.

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